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ORIGINS OF ORGANIC MATTER IN METEORITES

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Abstract: Several abiotic chemical processes acting in several different astrophysical and cosmogonic environments have contributed to the population of organic compounds preserved in carbonaceous meteorites. Those environments include interstellar molecular clouds and the near-surface regions of asteroid-sized objects in the early solar system. There is little evidence for synthesis of organic compounds from the primordial solar-nebular gas.

1. Introduction

Since its discovery nearly two hundred years ago meteoritic organic matter has excited considerable interest, much of it focusing on the possibility of "life in space". Many analyses have yielded results apparently consistent with a biogenic origin, but recent studies have shown convincingly that those earlier results were due to terrestrial contamination and that, when carefully analysed, meteoritic organic matter reveals no evidence for biological activity. Interest in this material has remained high, however, because of the information that it can supply concerning a key step in the sequence of events leading to the origin of life, namely the abiotic chemical reactions that generated those organic molecules that are believed to have constituted the "primordial soup". On Earth, the evidence from that epoch has long since been obliterated.

Over 400 individual organic compounds have been identified in carbonaceous meteorites (*e.g.*, CRONIN *et al.*, 1988), and more undoubtedly exist below current detection limits. The observed molecular species account for 10 to 20% of the organic C in a typical carbonaceous meteorite, the balance consisting of a poorly characterised macromolecular complex resembling terrestrial kerogen. This kerogen-like material contains few decipherable clues to its origin, the elemental and isotopic record having apparently been obscured by mixing and/or reprocessing. Consequently, most evidence bearing on origins is derived from the study of characterisable molecular species. In general, the most reliable data have been obtained for the Murchison CM carbonaceous chondrite, which fell in 1969 and was promptly collected before serious terrestrial contamination had occurred. (For details of the classification of carbonaceous, and other, meteorites, *see* SEARS and DODD, 1988.)

Table 1 lists a dozen mechanisms that could plausibly have been implicated in organic synthesis in the early solar system, or before. (Note that carbonaceous meteorites are very old, with mineralisation and compaction taking place no more than a few Myr after formation of the earliest solids in the solar system (MACDOUGALL *et al.*, 1984).)

Table 1. Possible sources and processes responsible for production of meteoritic organic matter.

Ion-molecule reactions in interstellar clouds
Radiation chemistry in interstellar grain mantles
Condensation in stellar outflows
Equilibrium reactions in the solar nebula
Surface catalysis (Fischer-Tropsch) in the nebula
Kinetically controlled reactions in the nebula
Radiation chemistry (Miller-Urey) in the nebula
Photochemistry in nebula surface regions
Liquid-phase reactions on parent asteroid
Surface catalysis (Fischer-Tropsch) on asteroid
Radiation chemistry (Miller-Urey) in asteroid atmosphere
Thermal reprocessing during metamorphism

Table 2. Some observations of meteoritic organic matter that represent potential diagnostic indicators of sources and/or processes responsible for organic synthesis.

Structural diversity
Prevalence of branched over straight chains
Amino/hydroxy acid proportions
Deuterium enrichment of organic matter
C,H isotopic distribution within kerogen-like material
C isotope distribution in homologous series
Isotopic differences between oxidized and reduced species

Despite this plethora of candidate mechanisms, debate has traditionally focussed on just two possible syntheses: catalyzed hydrogenation of CO (Fischer-Tropsch mechanism) and chemical reactions induced by electric discharge (Miller-Urey mechanism). Recently, however, most workers have concluded that more than one mechanism was probably involved (*e.g.*, CRONIN *et al.*, 1988; ANDERS, 1991; WING and BADA, 1992). One reason for this conclusion is the paucity of clear signatures in the meteoritic record for individual processes. Diagnostically significant patterns may be found in the meteorite data but in most cases these have not been traced to specific processes. Table 2 lists some of what now appear to be the key diagnostic indicators in meteoritic organic matter, and in what follows we discuss these as potential constraints on mechanisms of organic synthesis in the early solar system, or in the interstellar environment that preceded formation of the solar nebula.

Because it seems likely that a number of different processes acting in a number of different environments have been responsible for the population of organic molecules in meteorites, we must consider the likelihood that a compound analysed today was manufactured from precursor compounds synthesised during an even earlier epoch. It is not always clear *a priori* whether any particular property, *e.g.*, a molecular configuration or an isotopic composition, reflects the final synthesis or production of precursor material. This can lead to some ambiguity in interpretation of the meteoritic record.

2. Molecular Observations of Organic Matter

Analyses of meteoritic organic matter can yield two different kinds of genetically useful information: molecular, *i.e.*, information about the types of chemical compound present and their abundances; and isotopic, *i.e.*, information about the proportions of the stable isotopes of the biogenic elements, mainly C, H and N, present in different organic fractions. We shall start with evidence stemming from molecular analyses.

2.1. Structural diversity

Most classes of organic compound in an uncontaminated meteorite exhibit complete structural diversity, *i.e.*, all isomers that are stable are present, for virtually all species above the detection limit (*e.g.*, CRONIN *et al.*, 1988; SHIMOYAMA and SHIGEMATSU, 1992). This includes the higher aliphatic hydrocarbons that were previously thought to show only limited structural diversity (*e.g.*, HAYATSU and ANDERS, 1981). However, CRONIN and PIZZARELLO (1991) showed that structural diversity for these compounds increased with increasing distance from the surface of a large Murchison sample, with the innermost sample, *i.e.*, the most free from terrestrial contamination, revealing essentially complete structural diversity. Purines and pyrimidines appear to exhibit limited structural diversity, though contamination may be a problem in those analyses (CRONIN *et al.*, 1988).

The significance of structural diversity is that it is difficult to reconcile with any kind of structurally selective synthesis, such as grain-mediated catalysis, *e.g.*, Fischer-Tropsch. Thus, the Fischer-Tropsch synthesis produces predominantly normal, straight-chain alkanes, followed by slightly branched alkanes and alkenes (*e.g.*, HAYATSU and ANDERS, 1981), whereas a Murchison interior sample yields mainly a structurally diverse population of branched, alkyl-substituted cycloalkanes (CRONIN and PIZZARELLO, 1991).

2.2. Prevalence of branched over straight-chain species

The significance of this observation, valid for all known species except the hydroxy acids (CRONIN *et al.*, 1988) and possibly monocarboxylic acids (SHIMOYAMA *et al.*, 1989), is similar to that of the previous one. Abundance of a species appears to be a function of thermodynamic stability independent of reaction pathway, *i.e.*, synthesis in most cases apparently involved random addition of C atoms. There was thus no selection for straight-chain species, as found in certain catalysed reactions, such as Fischer-Tropsch (*e.g.*, HAYATSU and ANDERS, 1981).

2.3. Amino/hydroxy acid proportions

WOLMAN *et al.* (1972) noted that the proportions of different amino acids produced during a Miller-Urey spark-discharge experiment resembled those found in the Murchison meteorite. They concluded that the experimental results reflected operation of a Strecker cyanohydrin reaction taking place in the ammonia-containing aqueous medium of the Miller-Urey experiment, using as starting material aldehydes and HCN produced by the spark discharge, Fig.1. This suggested that the Murchison amino acids also resulted from action of a Strecker-type synthesis upon precursor compounds, possibly synthesised in an electric discharge. Later, PELTZER *et al.* (1984) showed that

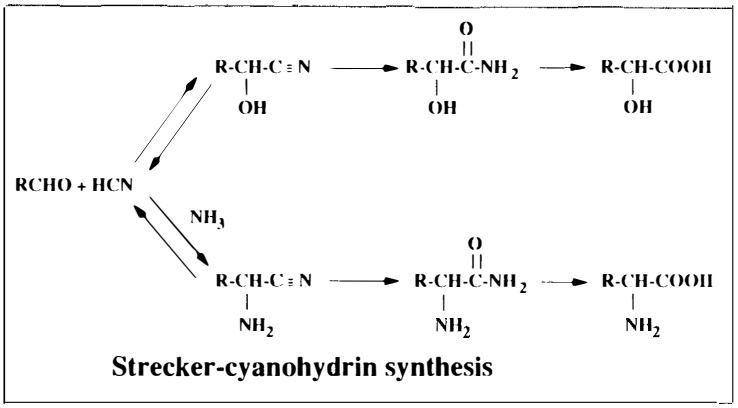

Fig. 1. Schematic representation of the pathways making up the Strecker cyanohydrin synthesis. This reaction scheme can take place in an aqueous medium containing aldehydes (and/or ketones), cyanide and ammonia. Note how the ammonia concentration controls the relative proportions of amino and hydroxy acids. After PELTZER *et al. (1984).*

for three out of four pairs of structurally similar amino and hydroxy acids in Murchison, the proportions of the amino to the hydroxy acid were consistent with synthesis by means of the Strecker cyanohydrin reaction in an aqueous medium characterised by a unique ammonium ion concentration. More recent results on a larger number of amino/hydroxy-acid pairs in Murchison (MILLER and BADA, 1992) reveal a wider range in apparent ammonia concentrations and therefore somewhat weaken the case for a Strecker-type synthesis. Clearly, further study of this reaction scheme is needed.

Whether or not the Strecker hypothesis is correct in detail, it is important to note that an aqueous environment is consistent with, indeed demanded by, the petrology of Murchison, and other carbonaceous meteorites (*e.g.*, KERRIDGE and BUNCH, 1979; MCSWEEN, 1979; BUNCH and CHANG, 1980). Furthermore, there is good reason to suppose that the reagents needed for the Strecker synthesis would have been present in the early solar system. Although production of ammonia in the solar nebula would have been kinetically inhibited, and aldehydes would not have been stable in a nebula of solar composition (FEGLEY and PRINN, 1989), those compounds and HCN are all known interstellar molecules (IRVINE and KNACKE, 1989) that could plausibly have survived formation of the solar nebula. These lines of evidence lead to a scenario in which amino, and probably other organic acids, were synthesised by reactions among interstellar molecules in the aqueous near-surface regions of an asteroid (BUNCH and CHANG, 1980; KERRIDGE, 1991).

3. Isotopic Systematics of Organic Matter

Molecular data of the kind described above have traditionally dominated the discussion concerning the origin of meteoritic organic matter. However, some of the most profound insights into this issue have come from the rapidly burgeoning field of organic isotope analysis. Figure 2 presents a broad overview of the C and H isotope systematics of organic components in Murchison, compared with the range of such systematics on Earth and with the composition inferred for the primordial solar nebula.

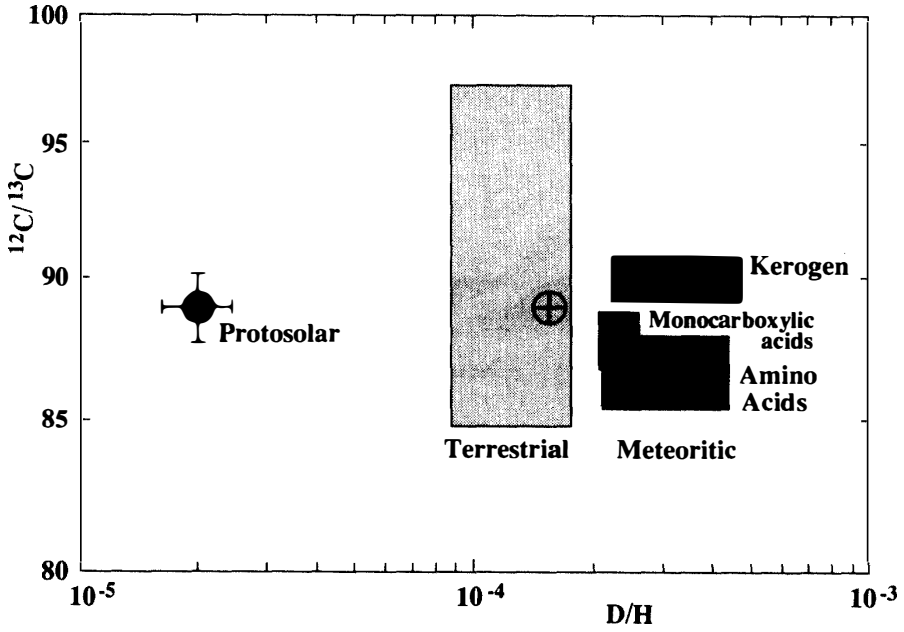

Fig. 2. Overview of C,H-isotope systematics in meteoritic organic components, compared with the protosolar values and the terrestrial range of values and mean composition. Note difference in scale between C and H data, and the very large enrichment of D in both terrestrial and meteoritic material.

Note the considerable difference in scale for the two sets of data; H shows vastly more variability than does C. Note also that the symbols for the organic fractions reflect the ranges of values observed within those fractions, not the uncertainty of the measurements. In what follows, we shall discuss the details of those intra-fraction variations, *i.e.*, examine the systematic patterns concealed within the symbols in Fig. 2.

3.1. Deuterium enrichment of organic matter

It is clear from Fig. 2 that the range of $\delta^{13}C$ values found in meteoritic organic matter lies within that observed on Earth and is consistent with known fractionation processes acting upon C with the isotopic composition characteristic of primordial solar material. However this is not the case for the H isotopes. All meteoritic organic species that have been analysed to date reveal considerable enrichment in deuterium including amino and carboxylic acids (EPSTEIN *et al.*, 1987; KRISHNAMURTHY *et al.* 1992), and kerogen-like material (ROBERT and EPSTEIN, 1982; KERRIDGE, 1983; KERRIDGE *et al.*, 1987). Figure 2 shows that these enrichments range up to a factor of three greater than the terrestrial D/H ratio, itself a factor of about eight enriched relative to the molecular hydrogen that dominated the primordial solar nebula. So great is this overall D enrichment that no known solar-system process is capable of generating it, Fig. 3 (GEISS and REEVES, 1981). Consequently, the meteoritic D enrichments are generally attributed to incorporation of molecules enriched in D by ion-molecule reactions in interstellar clouds prior to solar-system formation (*e.g.*, KOLODNY *et al.*, 1980; GEISS and REEVES, 1981; KERRIDGE, 1983). Note that it is the low temperatures of interstellar molecular clouds that are responsible for the high D enrichment, not some inherent property of

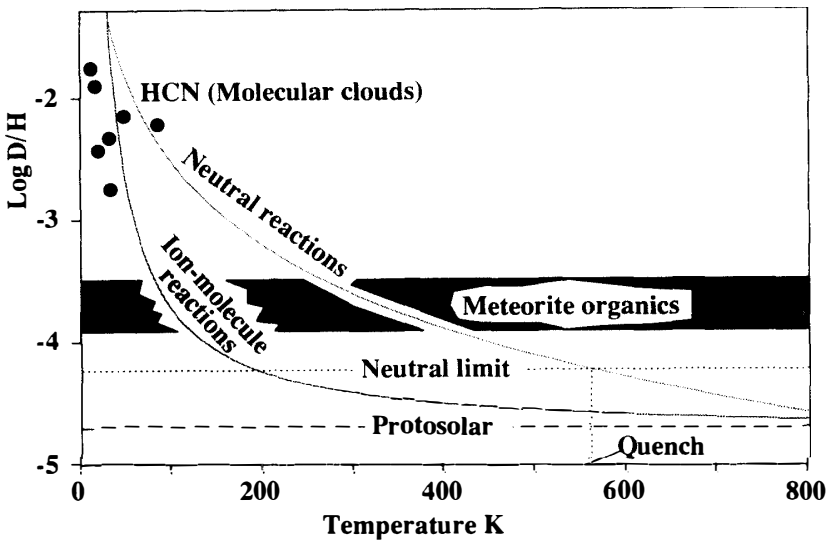

Fig. 3. Deuterium distribution in the early solar system. The two curves represent the temperature-dependent partition of D between protosolar H_2 and a typical organic molecule, the "neutral" curve corresponding to equilibriume exchange between neutral species, the "ion-molecule" curve to fractionation during a typical ion-molecule reaction. The point labelled "quench" represents the minimum temperature at which D would exchange between H_2 and CH_4 in a nebular gas of protosolar composition (GRINSPOON and LEWIS, 1987). The "neutral limit" is therefore the most D-rich composition that can result from equilibrium exchange in the primordial solar nebula. The points labelled HCN represent observations made in different interstellar molecular clouds.

the ion-molecule reaction schemes, which at any given temperature are actually less efficient than reactions among neutral species at fractionating isotopes.

Despite general agreement concerning the ultimate source of the deuterium enrichment, it is not known in what molecular form the D-enriched hydrogen entered the primitive solar system, though there are good reasons for believing that those presolar molecules included aliphatic hydrocarbons and aldehydes/ketones (KERRIDGE, 1991). Survival of such species poses strong constraints on the thermal history of the early solar system.

3.2. C,H isotopic distribution within kerogen-like material

The insoluble kerogen-like fraction is isotopically inhomogeneous, with both C and H exhibiting marked isotopic variations between the different molecular moieties analysed so far (KERRIDGE *et al.*, 1987). Those variations, in particular the $>6‰$ difference in $\delta^{13}C$ between aliphatic and aromatic C, Fig. 4, rule out production of the kerogen-like material by gas-phase reactions in the solar nebula, as proposed by MORGAN *et al.* (1991). The isotopic distribution in the kerogen-like material rather suggests production by degradation (possibly thermal or radiation-induced) of a melange of molecular species with a variety of origins.

It is tempting to try to relate the aromatic moieties in the kerogen-like material to the structurally very similar polycyclic aromatic hydrocarbons (PAH) found as molecular entities in carbonaceous meteorites (*e.g.*, HAHN *et al.*, 1988), however any genetic

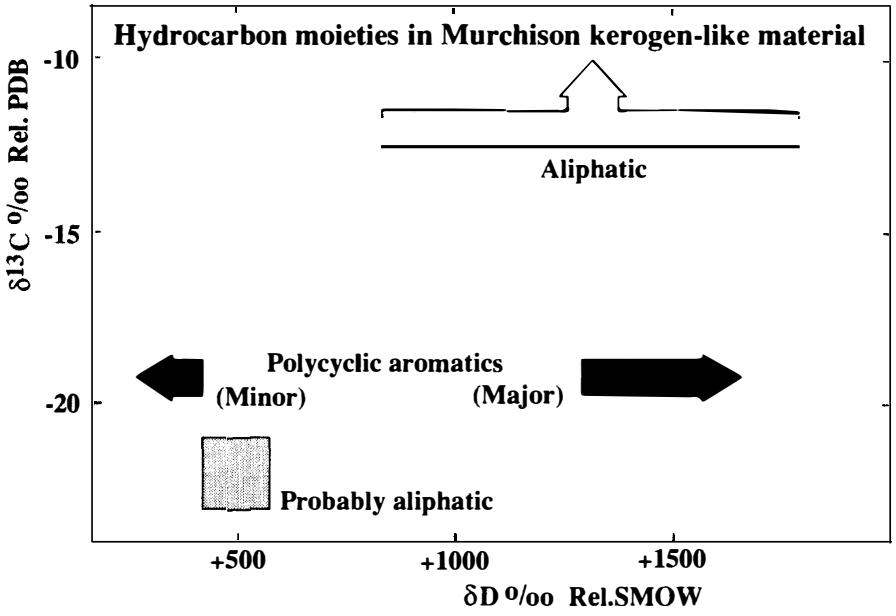

Fig. 4. Isotopic components identified during stepwise decomposition of kerogen-like material from the Murchison meteorite. None of the four components can be readily derived from any of the others. Isotope ratios expressed in the delta notation as deviations in parts per thousand relative to terrestrial standards. From KERRIDGE *et al. (1987).*

connection remains speculative at this time. Whether the individual PAH in meteorites could have formed by gas-phase reactions in the solar nebula (MORGAN *et al.*, 1991) is not known, but such an origin is problematical for the aromatic moieties in the kerogen-like material because of their high D contents. (The D contents of the individual PAH molecules in Murchison are not currently known.) This would require substantial local D enrichment of the nebular H_2 gas, which is most unlikely but which cannot be ruled out at this time.

WING and BADA (1992) have pointed out that the high proportion of unalkylated to alkylated PAH in Murchison indicates a high formation temperature, in the range 400 to 800°C, consistent with production in stellar outflows, *i.e.*, a presolar origin (ALLAMANDOLA *et al.*, 1987).

3.3. C isotope distribution in homologous series

YUEN *et al.* (1984) showed that the homologous series of both the light alkanes and the monocarboxylic acids exhibited similar systematic decreases of $^{13}C/^{12}C$ with increasing C number, at least up to C_5, Fig. 5. The carboxylic-acid pattern is probably carried by the aliphatic C, with the carboxyl C maintaining approximately the isotopic composition of CO_2 in the meteorite. YUEN *et al.* argued that the C isotopic pattern was inconsistent with production of the light alkanes by "cracking" of higher hydrocarbons but pointed instead towards build-up of the homologous series by progressive addition of C atoms. The data also suggested a close relationship between the light alkanes and the aliphatic precursors to the carboxylic acids (YUEN *et al.*, 1984), a connection which might well be extended to include the amino acids as well (SHIMOYAMA

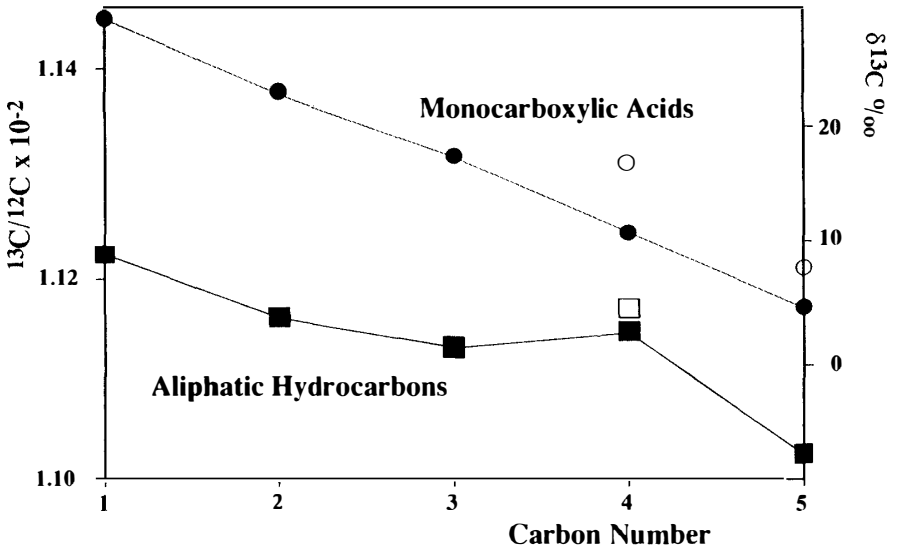

Fig. 5. $^{13}C/^{12}C$ *ratios for individual aliphatic hydrocarbons and carboxylic acids extracted from the Murchison meteorite, plotted against C number. For example, 1 denotes methane or* CO_2*, 2 ethane or acetic acid, and so forth. Values for branched isomers are indicated by open symbols. After* YUEN *et al. (1984).*

et al., 1986; KERRIDGE, 1991).

3.4. Isotopic differences between oxidized and reduced species

Inspection of Figs. 2, 4 and 5 reveals that oxidized species, *e.g.*, amino and carboxylic acids, are systematically enriched in ^{13}C compared with reduced species, *e.g.*, alkanes and kerogen-like material (CHANG *et al.*, 1978). Although this has been attributed to kinetic isotope fractionation during operation of the Fischer-Tropsch synthesis (HAYATSU and ANDERS, 1981), more recent laboratory simulations (KERRIDGE *et al.*, 1989; YUEN *et al.*, 1990) indicate that this is unlikely. The cause of this isotopic difference is not understood in detail.

It is also worth noting that simulations of several reaction schemes, including Fischer-Tropsch and Miller-Urey, have failed to reproduce the variability found for H and N isotopic compositions in meteoritic organic matter (KUNG *et al.*, 1979; KERRIDGE *et al.*, 1989). (Nitrogen isotopic analysis of various organic fractions of Murchison has yielded $\delta^{15}N$ values spanning a range of approximately 100‰ (*e.g.*, BECKER and EPSTEIN, 1982; KERRIDGE *et al.*, 1987; EPSTEIN *et al.*, 1987)).

4. Summay

The foregoing considerations strongly suggest production of meteoritic organic compounds and their precursor materials by a variety of mechanisms in a variety of environments. There is good evidence that those environments included dense molecular clouds and the surface regions of asteroid-sized objects. On the other hand, there is presently no compelling evidence for organic synthesis in the solar nebula, including the frequently cited Fischer-Tropsch-type reaction. At present, the most suggestive

meteoritic evidence for operation of a catalysed synthesis such as Fischer-Tropsch is the observation of carbonaceous coatings on certain (possibly catalytic) mineral grains in some carbonaceous chondrites (ALPERN and BENKHEIRI, 1973). However, it is difficult to exclude the possibility that the carbonaceous material was synthesised elsewhere and was subsequently picked up by the mineral grains during regolith processing on the parent asteroid.

Obviously, the lack of firm evidence for organic synthesis in the nebula should not be taken as evidence against such a hypothesis, however it does limit seriously the information that we can derive from meteorites concerning carbon chemistry in the solar nebula. At present, the meteoritic organic record may be more informative about physical conditions in the nebula, which apparently permitted survival of interstellar methane and other light alkanes (as well as a variety of less volatile species), possibly trapped in icy grain mantles (*e.g.*, GREENBERG 1982).

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